

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

VINCENT J. GATTO, ET AL.)	
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APPLN NO.: 10/788,850)	
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)	EXAMINER: TAYLOR V. OH
CONFIRMATION NO.: 6851)	
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CUSTOMER NO.: 65895)	
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PREPARATION OF STERICALLY HINDERED)	
HYDROXYPHENYLCARBOXYLIC ACID ESTERS)	
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Commissioner for Patents
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Sir:

DECLARATION UNDER 37 C.F.R. §1.132

Dr. Mahmood Sabahi hereby declares as follows:

1. I am a chemist employed by Albemarle Corporation, the assignee of the above-identified application by recorded assignment.
2. In 1979, I received the degree of Ph.D. in chemistry from Syracuse University.
3. Since 1990, I have been continuously employed as a chemist by Albemarle Corporation and the predecessor thereof, Ethyl Corporation, at their research laboratory facilities in Baton Rouge, Louisiana.
4. The test work described herein was conducted by me at the Albemarle Process Development Center in Baton Rouge, Louisiana.
5. In the first part of this work, Example 1 of Haeberli et al. (U.S. 4,228,297) was repeated with some small changes, and by performing an analogous experiment according to the present claims.
6. In the procedure following Haeberli et al., a dry 500 mL three-necked flask was equipped with a condenser, a calibrated dropping funnel, a stirrer, and a nitrogen inlet. The flask

was purged with nitrogen and 2,6-di-tert-butylphenol (103 g) was added and heated to 70°C. Then potassium t-butoxide (1.4 g) was added, followed by isopropyl alcohol (2 mL), and the resulting mixture was heated to 107 to 110°C. Methyl acrylate (47.3 g) was placed in the addition funnel and added to reaction mixture over a period of two hours at a temperature range of 107-110°C. The resulting mixture was heated at this temperature range for another three hours. Excess methyl acrylate was distilled under vacuum (20 mmHg and 110°C) and the reaction mixture was cooled to 70°C, at which temperature isooctanol (78 g) and lithium hydroxide (0.47 g) were added while keeping the reaction mixture under a constant flow of nitrogen. Vacuum was applied to the flask (20 mmHg) and the contents were heated to 140-145°C over two hours and heated for another three hours at this temperature while collecting methanol. The vacuum was released with nitrogen, the reaction mass was cooled to 70°C, and glacial acetic acid (3.0 g) was added and the mixture was stirred for 30 minutes. Ethanol (132 g) was added and the resulting mixture was clarified over a Celite bed (25 g). The filtrate was cooled to room temperature. The product did not precipitate out of solution, so the ethanol was removed under vacuum at 70°C. The product, an oil, was analyzed by inductively-coupled plasma mass spectrometry (ICP-MS) for potassium, lithium, and phosphorus content; results are shown in Table 1 below.

7. In the experiment according to the present claims, a dry 500 mL three-necked flask was equipped with a condenser, a Dean-Stark trap, a stirrer, and a nitrogen inlet. The flask was purged with nitrogen and 2,6-di-tert-butylphenol (103 g), potassium hydroxide (0.65 g), dimethyl sulfoxide (2 mL), and toluene (40 g) were added and heated to 140°C and water was collected in the trap. After complete removal of water from the reaction mixture, the Dean-Stark trap was replaced with a calibrated addition funnel and methyl acrylate (50 g) was added at 120°C over about 40 minutes. The resulting mixture was heated at this temperature for an additional three hours. Excess methyl acrylate was removed under vacuum (20 mmHg) at 120°C. Then isooctanol (78 g) was added and vacuum was applied (20 mmHg) at a temperature range of 150 to 155°C and methanol was collected until the conversion was complete. The reaction mixture was cooled to about 80°C and phosphoric acid solution (prepared by mixing 1 g of 85% H₃PO₄ with 4 g

of water) was added under nitrogen. The resulting mixture was heated to 140°C and kept at this temperature with stirring for one hour. Vacuum (2-4 mmHg) was applied, and the reaction mixture was heated to 150°C to remove excess isooctanol, water, and other volatiles. As the volatiles are removed, the liquid becomes cloudy due to the precipitation of the phosphate salts as fine particles, which remain suspended in the liquid. The resulting cloudy oil was cooled to about 100°C and filtered through a short Celite bed (about 25 g), removing the precipitated phosphate salts; a clear oil was obtained as the filtrate. The clear oil was analyzed by inductively-coupled plasma mass spectrometry (ICP-MS) for potassium and phosphorus content; results are shown below in Table 1.

8. The results of the analyses of the products of the two experiments are shown in Table 1.

TABLE 1

Element	Haeberli et al.	Present case
Potassium	2172 ppm	183 ppm
Phosphorus	N/A	23 ppm
Lithium	489 ppm	N/A

9. In addition, it is noted that a commercially-produced product using the above process in accordance with the present claims in a four-month period had an average potassium content of 14 ppm.
10. Upon cooling to room temperature, the product of the Haeberli et al. procedure described above became cloudy, and a second lower phase separated with a clear orange-red oil on top (see Figure 1 below). Figure 1 is a photograph of this solution from the Haeberli et al. procedure. It appears that the product from Haeberli et al. contains a large amount of potassium acetate that precipitates out of the oil on standing.
11. The oil from the experiment according to the present claims remained clear on standing at room temperature for five days (see Figure 2 below). Figure 2 is a photograph of this solution of the experiment according to the present claims, alongside the precipitate-containing solution from the Haeberli et al. procedure.

12. It is my considered opinion that the presently claimed process provides a much better separation of the product from the salt produced in the neutralization reaction than the process described in Haeberli et al., based on the above results.



Figure 2



Figure 1

13. In the second part of this work, a calculation was performed to estimate the economic advantages of the present claims over the process in Haeberli et al.
14. For the calculation, phosphoric acid was replaced with acetic acid as described in Example 7 of the present application (Specification, Pages 49-50); the calculations accounted for water washes as in Example 7. Stripping and drying time for the final product were estimated from the cost of stripping and drying in the present process, multiplied to account for the larger amount of water used in the water washes of the acetic acid route as in Example 7. The cost of making a product in this manner with acetic acid was calculated and compared to the cost of producing a product using phosphoric acid.
15. From the calculation, the cost of one kilogram of product from the phosphoric acid process of the invention is 12.4% less than a process using the acetic acid/water wash. In other words, elimination of water washing and subsequent drying (by using phosphoric acid) reduces the cost of production by 12.4%.
16. It is my considered opinion that one of the economic advantages of the of the present claims is the 12.4% reduction in cost for producing a product using phosphoric acid, based on the above calculation. In addition, the claimed process is environmentally advantageous because it eliminates hazardous waste water that results from the water washes that are necessary when acetic acid is used. Thus, another economic advantage of the presently claimed process is the avoidance of disposal costs for this waste water.
17. I hereby declare that all statements made herein of my own knowledge are true and that all statement made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: August 12, 2009

Mahmood Sabahi
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